

Appl. No. 10/619,698

Response dated October 26, 2005

Reply to Office Action of September 28, 2005

Amendments to the Specification

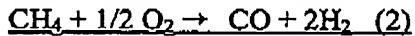
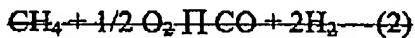
Please replace specific paragraphs [0005], [0008], [0020], [0021], [0038], [0042], and [0047]-[0050] of the specification with the paragraphs set out below, all cited paragraph numbers originating from the specification as filed.

[0005] Current industrial use of methane as a chemical feedstock proceeds by the initial conversion of methane to carbon monoxide and hydrogen by either steam reforming, which is the most widespread process, or by dry reforming or by autothermal reforming. Steam reforming currently is the major process used commercially for the conversion of methane to synthesis gas, proceeding according to Equation 1.



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[0008] The catalytic partial oxidation ("CPOX") or direct partial oxidation of hydrocarbons (e.g., natural gas or methane) to syngas has also been described in the literature. In catalytic partial oxidation, natural gas is mixed with air, oxygen-enriched air, or oxygen, and introduced to a catalyst at elevated temperature and pressure. The partial oxidation of methane yields a syngas mixture with a H₂:CO ratio of 2:1, as shown in Equation 2.



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[0020] Under autothermal process conditions there is no external heat source required to the partial oxidation reaction. The net partial oxidation reaction conditions are promoted by optimizing the concentrations of hydrocarbon and O₂O₂ in the reactant gas mixture, preferably with a mole ratio in the range of from about 1.5:1 to about 3.3:1 carbon:O₂O₂. In some embodiments, steam may also be added to produce extra hydrogen and to control (i.e., reduce) the outlet temperature. The steam can be added as an additional flow stream in the gas

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distributor, can be premixed with one of the feed gases upstream of the gas distributor, or can be added separately, as desired. Regardless, it is preferred that the ratio of steam to carbon (by weight) range from 0 to 3.

[0021] Pressure, residence time, amount of feed preheat and amount of nitrogen dilution, if used, also affect the reaction products. The process also preferably includes maintaining a catalyst residence time of no more than about 100 milliseconds for the reactant gas mixture and in some reactor schemes even less. This is accomplished by passing the reactant gas mixture over, or through the porous structure of the catalyst system at a gas hourly space velocity of about 20,000 - 100,000,000 h^{-1} , preferably about 100,000 - 25,000,000 h^{-1} . In preferred embodiments of the process, the catalyst system catalyzes the net partial oxidation of at least 80% of a methane feedstock to CO and H_2H_2 with a selectivity for CO and H_2H_2 products of at least about 80% CO and 80% H_2H_2 .

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[0038] The number of fluid outlets is preferably high relative to the total surface area of the catalyst. For example, it may be preferred to provide at least 7 outlets, preferably evenly spaced across the cross-sectional area of the reactor or catalyst bed. In some embodiments, the number of outlets per square foot is preferably at least 1, and more preferably at least 25. Still other embodiments may have as many as 100, 1000, 2,000, or 6,000 outlets per ft^2 , or more. If the reactant gases are kept separate as they pass through the gas distributor, the number, size and proximity of the outlets is preferably such that the fluids are quickly uniformly dispersed into each other upon exiting the gas distributor block. This can be achieved by having at least 20 outlets of each gas per square foot.

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[0042] A preferred process for producing synthesis gas employs a monolithic or divided catalyst containing a catalyst composition that is effective for catalyzing the conversion of methane or natural gas and molecular oxygen to primarily CO and H_2H_2 by a net catalytic partial oxidation (CPOX) reaction. Preferably employing a fast contact (i.e., millisecond range)/fast quench (i.e., less than one second) reactor assembly, a feed stream comprising a light hydrocarbon and an oxygen-containing gas is contacted with the catalyst. The reactor is preferably operated at a

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reactant gas pressure greater than 1 atmosphere (>100 kPa), more preferably above 2 atmospheres, which is advantageous for optimizing syngas production space-time yields. One suitable reaction regime is a fixed bed reaction regime, in which the catalyst is retained within a reaction zone in a fixed arrangement, as conceptually illustrated in Figure 4.

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[0047] In operation, a stream of light hydrocarbon, such as methane, or other feed gas is fed into feed injection opening 212. An oxygen-containing gas, such as air or oxygen is fed into a second injection opening 214, which is preferably positioned close to catalyst 224. It should be understood that the feed injection openings in the reactor, the manifold(s) and the micro-channel gas distributor 218 can be configured differently from the configuration shown in Figure 4 without affecting the principles or operation of the process. For example, as discussed above and illustrated in Figure 2, the flow channels in gas distributor 18 could be configured such that the oxygen is kept separate from the methane and is not mixed with the light hydrocarbon until it exits the gas distributor. In this embodiment, mixing of the gases occurs immediately before or during the contacting of the feed gas stream with a hot catalyst. Such configurations may help reduce the occurrence of unwanted side reactions that might otherwise rapidly occur during or after mixing of $\Theta 2O_2$ with the hydrocarbon components but prior to contacting the catalytic surfaces of the reaction zone.

[0048] Air or a mixture of air and oxygen can be substituted for the pure oxygen. However, since the presence of $N2N_2$ in the reactant gas mixture can be problematic (e.g., forming unwanted nitrogen-containing compounds), it is preferable in most cases to use pure oxygen instead of air. The hydrocarbon feedstock may be any gaseous hydrocarbon having a low boiling point, such as methane, natural gas, associated gas, or other sources of light hydrocarbons or alkanes having from 1 to 5 carbon atoms. The hydrocarbon feedstock may be a gas arising from naturally occurring reserves of methane. Preferably, the feed comprises at least about 80% by volume methane. The hydrocarbon feedstock may also include some steam and/or $CO2CO_2$, as sometimes occurs in natural gas deposits.

[0049] The methane-containing feed and the $\Theta 2O_2$ -containing feed are mixed in such amounts as to give a carbon (i.e., carbon in methane) to oxygen (i.e., molecular oxygen) molar ratio from about 1.5:1 to about 3.3:1, more preferably, from about 1.7:1 to about 2.1:1. The stoichiometric molar

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ratio of about 2:1 (CH_4 : O_2O_2) is especially desirable in obtaining the net partial oxidation reaction products ratio of 2:1 H_2H_2 :CO. As mentioned above, the hydrocarbon or reactant gas mixture is preferably preheated to about 30°C - 750°C before contacting the catalyst.

[0050] During mixing, the feed gases are shielded by radiation barrier 222 from radiant heat that is generated downstream in the process. It is preferred that the temperature on the upstream side of barrier 222 be in the range of about 30°C to about 500°C, preferably no more than about 750°C, to help initiate the CPOX reaction. Excessive preheating the feed gases can cause unwanted homogeneous reactions to occur that reduce the selectivity of the process for the desired CO and H_2H_2 products. In some instances, it may also be desirable to briefly supplement the hydrocarbon feed with propane or another pure hydrocarbon to facilitate rapid initiation of the CPOX reaction. After the gases pass barrier 222, they flow past catalytic device 224 and are simultaneously heated to 350°C - 2,000°C, preferably not exceeding 1,500°C, and more preferably staying in the range of about 400°C to about 1,200°C.